

The role of sidestream recycle in hydrogen isotope separation and column cascade design

Robert H. Sherman ^a, David J. Taylor ^a, T. Yamanishi ^b, M. Enoeda ^b, S. Konishi ^b,
K. Okuno ^b

^a *Tritium Systems Test Assembly, Los Alamos National Laboratory MS-C348, PO Box 1663, Los Alamos, NM 87545, USA*

^b *Japan Atomic Energy Research Institute, Tokai-mura, Ibaraki-ken, Japan*

Abstract

Sidestream recycle combined with sidestream equilibration is important in hydrogen isotopic distillation processes because it offers a means to reduce the number of columns required for the extraction of pure homonuclear species. This directly implies simpler systems, reduced control problems, and reduced material inventories. Measurements were recently completed for a single distillation column using feed compositions (about 50:50 D:T) and product flows similar to those expected in an ITER-type device with recycle of an equilibrated sidestream withdrawn from the column. Dynamic studies were conducted with flowrates changing as might be expected for typical Tokamak operations. These experimental results are compared with computer simulations of the dynamic process. The impact of these sidestream recycle studies on the design of isotope separation systems is discussed, especially with respect to column design, tritium inventory, dynamic performance, stability, and system control.

1. Introduction

Distillation is often the most efficient means to separate the components of a mixture. A two-component mixture can be separated into product streams of high purity using a single distillation column. However, separation of hydrogen isotopes (e.g. D_2/T_2) by distillation is complicated by the existence of stable mixed-isotope molecules (molecular DT), so that a sequence of distillation columns and equilibrators is generally required when the feed tritium is primarily DT. The technique of sidestream recycle with sidestream equilibration can substantially enhance the purity of at least one of the product streams from a single-column isotope separa-

tion system (ISS) by strategically withdrawing a sidestream containing relatively high concentrations of the mixed-isotope molecule, which is broken up by equilibration. In a previous paper [1] data was presented for a single-column distillation experiment on an equilibrated mixture of essentially H_2 and D_2 . After a period of normal column operation, a sidestream was withdrawn, isotopically equilibrated, and re-injected into the column. The performance was then measured using Raman spectroscopy for dynamic compositional analyses, demonstrating an improvement in reboiler product purity from 55% to 99% using sidestream recycle. A second experiment was also performed using a feed composition H:D:T of 0.204:0.6581:0.115. In the cur-

rent study, an additional mixture was studied which contained significantly higher concentrations of T_2 , more closely resembling feeds that might be encountered in processing Tokamak exhaust gas.

2. Experimental details

The first column of the Tritium Systems Test Assembly (TSTA) 4-column ISS was chosen for this study, as for the previous study, because it has 83 theoretical stages, 3 alternative feed-injection points, and 7 sample withdrawal locations. The feed stream was injected into the 29th stage (counted from the reboiler). A side-stream was extracted via a metal-bellow pump from the 9th stage, equilibrated, and combined with the main feed for re-injection into the column. The nominal external feed composition H:D:T to the first column was 0.069:0.301:0.630 at a rate of 1319 sccm during the sidestream recycle experiments. The column pressure was 808 Torr; reflux ratio 120; condenser and reboiler flow rates 415 and 904 sccm respectively; recycle flow rate 4112 sccm; and the reboiler contained 45 ml of liquid. The reboiler composition was monitored every 3 min over a period of 8 h using on-line Raman spectroscopy of the hydrogen isotope rotational lines. A comparison of the experimental observations of reboiler composition with computer simulations is shown in Fig. 1. Note that the experimental steady-state value of tritium purity agreed well with the predicted value from computer simulations, and demonstrated an improvement in tritium purity from 80% to 95% using side-stream recycle. While the observed dynamics initially agreed with the simulation, the column achieved its high steady-state purity within about 3 h, substantially faster than predicted by the simulation. This may be due to limitations of the present Raman spectroscopic system which does not permit simultaneous observation of more than one sample tap.

Further analysis of the data presented in the previous paper [1] results in a value of the liquid holdup on the packing of 10.1% of the superficial volume for DT and T_2 mixtures. This compares favorably with the previously measured value of 9.5% for H_2 and D_2 [2,3].

3. Design

In designing a system of columns for the decomposition of DT and the extraction of T_2 there are choices of either (i) a two-column cascade with recycle from the second column back to the first, or (ii) a single column

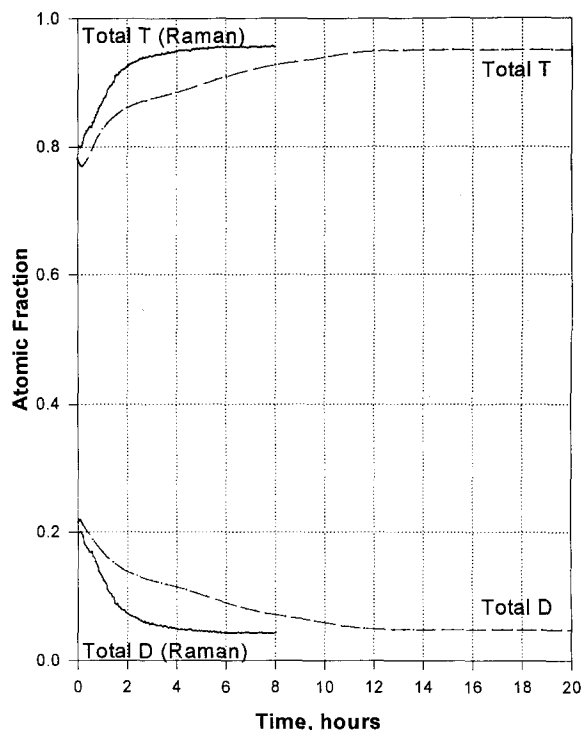


Fig. 1. Comparison of observed (solid lines) and calculated (dashed lines) reboiler composition following initiation of sidestream recycle.

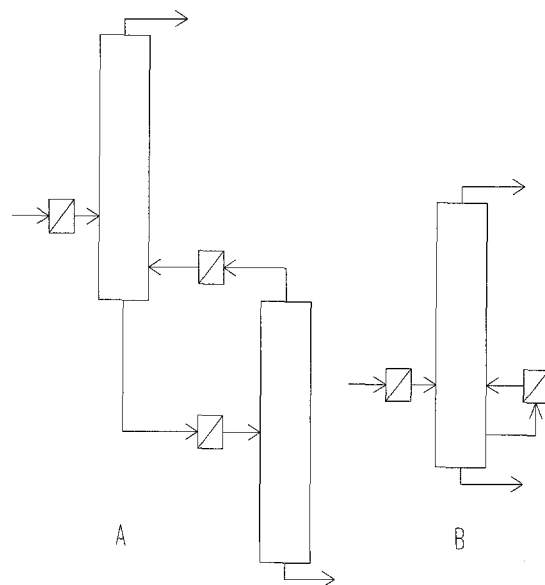
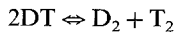
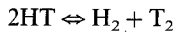


Fig. 2. Two- and one-column configurations showing distillation columns and equilibrators.

with sidestream recycle; these are shown in Fig. 2. Sulzer Brothers has designed a system of two columns for the recovery of T_2 from the moderator of the ILL reactor [4,5]. The final column contains an intermediate reboiler with a passive recycle stage, i.e. a sidestream is withdrawn into a volume which contains no catalyst. Gas movement is effected by alternately cooling and heating this external volume and the isotopic equilibrium occurs due to β -induced exchange. The Los Alamos distillation dynamic computer simulation code has been used to determine optimal conditions for both choices of column configuration assuming a feed of 100 mol h^{-1} of $H:D:T = 0.01:0.495:0.495$, similar to what might be expected from the processing of the high-tritium fuel stream from a Tokamak fusion energy device. In this case the DT molecule must be decomposed as given by the equilibrium reaction



A second set of conditions was determined for a feed stream of $H:D:T = 0.495:0.01:0.495$ such as might be encountered in blanket or waste processing. In this case it is the HT molecule which is to be removed according to



Operating conditions are summarized in Table 1. Fig. 3 shows the results of changing the sidestream location for the single-column sidestream recycle case. In a multicomponent distillation one or more of the components (such as HT or DT) may reach a maximum concentration at some intermediate stage. The location of such a maximum is often very dependent on the ratio of top to bottom flows. As the reboiler withdrawal fraction changes the maximum will move up or down the column, and column inventory may show large

Table 1
Column parameters

Parameter	Diatomic species	
	HT	DT
Stages	83	83
Pressure	760	760
Reflux ratio	20	20
Top fraction	0.51	0.49
Bottom fraction	0.49	0.51
Sidestream rate, relative to feed	6:1	6:1
Optimal withdrawal stage	10	50

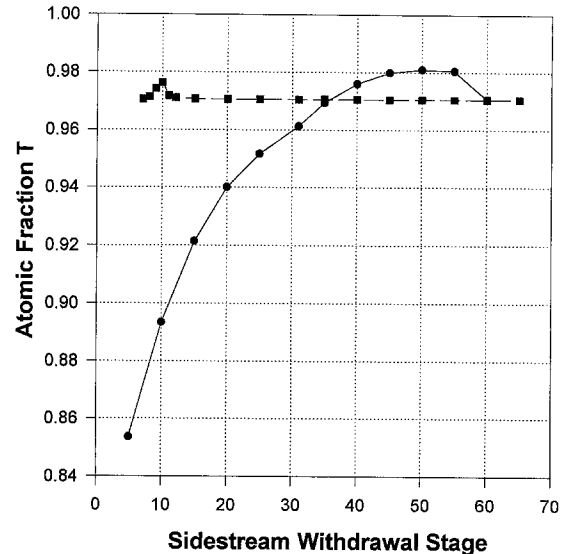


Fig. 3. Effect of location for sidestream withdrawal on reboiler purity: ■, HT; ●, DT.

changes. The optimal location of a sidestream stage is near this maximum; however the extraction of the sidestream will, in turn, alter the column concentration profiles. Once the optimal location for sidestream withdrawal is determined one can then decide the optimal location for re-injection of the equilibrated recycle stream. Such a determination is shown in Fig. 4 from

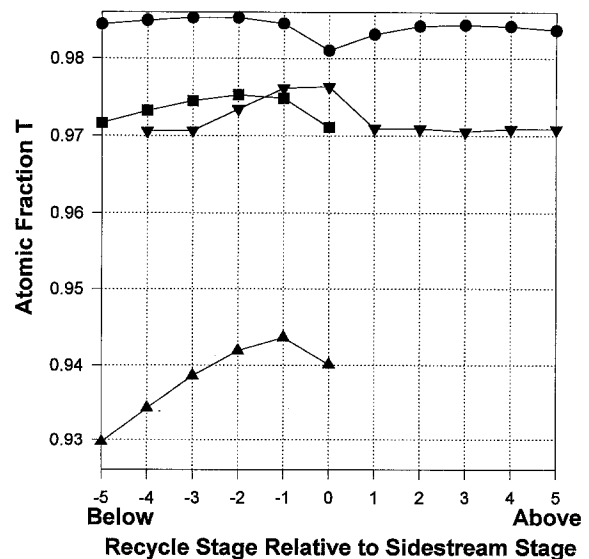


Fig. 4. Effect of stage offset in re-injection of equilibrated sidestream on reboiler purity. Sidestream withdrawal at stage: ●, 50 (DT); ■, 36 (DT); ▲, 20 (DT); ▼, 10 (HT).

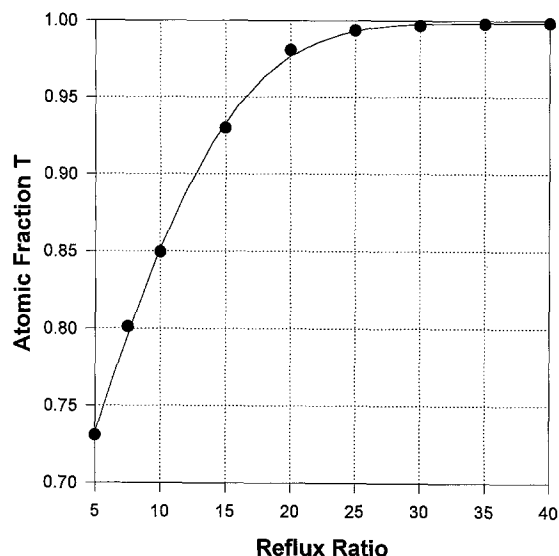


Fig. 5. Effect of reflux ratio on reboiler purity for the DT case.

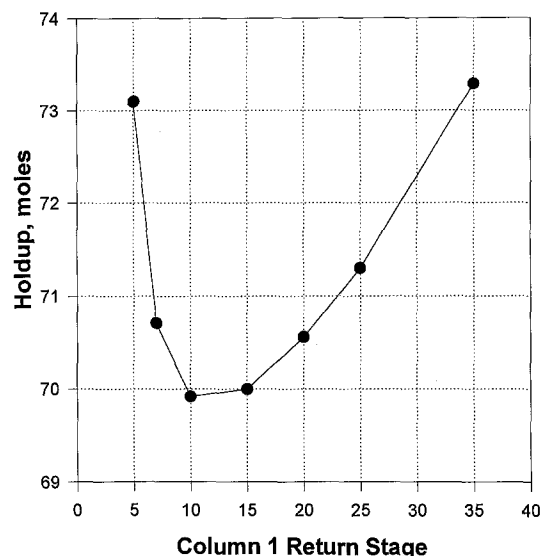


Fig. 7. Effect of return stage on holdup for two columns with DT feed.

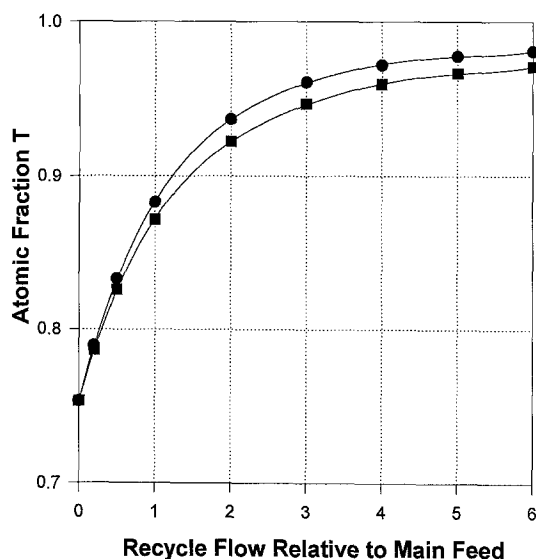


Fig. 6. Effect of sidestream flow rate relative to main feed flow rate for the DT case. Sidestream at stage: ●, 50; ■, 36.

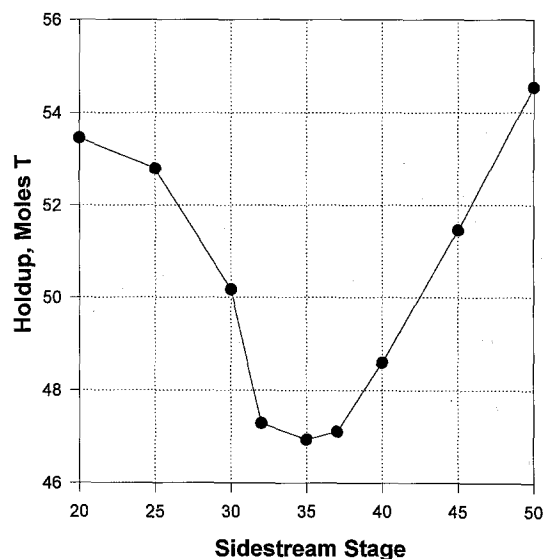


Fig. 8. Effect of sidestream stage on holdup for single column with DT feed.

which it is seen that the optimal return location is generally one or two stages below the sidestream extraction stage. Two other important parameters in the overall column design are the reflux ratio and the sidestream flowrate relative to the main feed rate; these are shown in Figs. 5 and 6. It can be seen that a return flowrate factor of at least 3 is necessary for effective application of sidestream recycle. The effect of return

stream location in the two-column configuration is shown in Fig. 7; the effect of sidestream location on tritium holdup is shown in Fig. 8. These figures make clear the desirability of being able to adjust sidestream and recycle taps. The percentage effect on column inventory is much stronger in the sidestream recycle case.

Table 2
Column parameters for holdup comparison

Parameter	Two-column	Two-column	Single-column
	1	2	
Stages	83	65	83
Pressure (Torr)	760	760	760
Diameter (cm)	8.5	8.1	9.5
Reflux ratio	20	5	25
Top fraction	0.495		0.495
Bottom fraction		0.505	0.505
Reboiler power (W)	235	163	317
Feed stage	29	45	29
Feed rate (mol h ⁻¹)	100	200.5	100
Sidestream stage			35
Recycle stage	10		33
Recycle rate (mol h ⁻¹)		150	600
Fraction T in distillate	7.7×10^{-3}		7.60×10^{-3}
Fraction T in reboiler		0.9708	0.9726
Reboiler holdup (mol)	0.2	0.2	0.2
Total holdup: moles H ₂	0.34	0	0.33
Total holdup: moles D ₂	85.4	22.9	78.3
Total holdup: moles T ₂	16.8	53.1	46.9

It is always simpler to control a single column. Each additional column in a cascade adds additional complication, especially if there are recycle streams to a previous column. The choice of a one- or two-column system is often a quite subjective decision. A two-column system offers greater flexibility in its ability to accommodate variations in the feed composition. To achieve minimum inventory the one-column system is preferable. A method of accommodating variations in feed composition is to provide a series of withdrawal/re-injection taps which could be selected on-line to provide optimal column operation.

The important parameters are summarized in Table 2. From the total T₂ holdup it can be seen that a

Table 3
Comparison of two- and one-column systems

Parameter	Two-column	Single-column
Number of stages	More	Less
Reboiler power	More	Less
Holdup	More	Less
Ease of control	Complicated	Simple
Flexibility	More	Less

two-column system will have a holdup of 69.9 mol as compared with 46.9 mol for a single column with side-stream recycle, corresponding to at least 1.5 times higher tritium inventory.

As mentioned above, various tradeoffs must be weighed in reaching the final design of a system of columns to perform a specified separation. These tradeoffs are summarized in Table 3.

4. Conclusions

Further verification of the application of the side-stream recycle technique to hydrogen isotope separation has been carried out. Traditionally it has been necessary to make use of a two-column cascade to extract a pure T₂ product from a D₂–DT–T₂ mixture. Each of the two columns would have a significant inventory of tritium. The use of the sidestream recycle configuration permits the elimination of one distillation column and can result in a tritium inventory reduction by a factor of 1.5. Additionally, control of a single column is simpler than a two-column cascade.

Acknowledgment

This work was supported by the US DOE Office of Fusion Energy and the Japan Atomic Energy Research Institute.

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